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Magnetic Anisotropy in the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ MAX Phase

Ruslan Salikhov^{a,*}, Anna S. Semisalova^{a,b,c}, Andrejs Petruhins^d, Arni Sigurdur Ingason^d,
Johanna Rosen^d, Ulf Wiedwald^a and Michael Farle^a

^aFakultät für Physik and Center for Nanointegration (CENIDE), Universität Duisburg-Essen, Duisburg 47057, Germany; ^bFaculty of Physics, Lomonosov Moscow State University, Moscow 119991, Russia; ^cInstitute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Dresden D-01328, Germany;

^dThin Film Physics Division, Department of Physics, Chemistry and Biology (IFM), Linköping University, Linköping SE-581 83, Sweden

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Magnetic MAX phase $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ thin films grown epitaxially on $\text{MgO}(111)$ substrates were studied by ferromagnetic resonance at temperatures between 110 and 300 K. The spectroscopic splitting factor $g = 2.00 \pm 0.01$ measured at all temperatures indicates pure spin magnetism in the sample. At all temperatures we find the magnetocrystalline anisotropy energy to be negligible which is in agreement with the identified pure spin magnetism.

Keywords: Magnetic MAX Phase, Ferromagnetic Resonance, g -Factor, Magnetic Anisotropy

MAX phase materials based on nanolaminated ternary compounds have been found to show unique and intriguing properties ranging from spectacular mechanical to self-healing and superconducting properties.[1–3] MAX phases $(\text{M}_{n+1}\text{AX}_n)$, where $n = 1, 2$, or 3 are layered hexagonal structures, where M is an early transition metal (Sc, Ti, V, Cr, Zr, Nb, Mo, Hf, or Ta), A is an A-group element (e.g. Al, Ga, or Ge), and X is either C or N. Recently, Mn- and Cr-based MAX phases have attracted considerable interest due to their magnetic properties. Density functional theory calculations have predicted antiferromagnetic (AFM) long-range order in Cr_2AC (where $A = \text{Al}$ or Ge) structures,[4,5] while ferromagnetic (FM) ordered Cr moments in an external magnetic field below 80 K [6,7] have been suggested from experiments. On the other hand, more recent work has shown that the Invar-like behavior discussed in Jaouen et al. [7] is quite common in similar structures, and probably cannot be attributed to the magnetic behavior.[8] Another experimental work suggested that the Cr_2GeC MAX phase can be classified as a correlated-electron paramagnetic metal in the vicinity of an FM quantum critical point.[9] The possibility to achieve a stable FM order at room temperature in nanolaminated

MAX phases in bulk or thin film form makes these systems extremely interesting for both fundamental materials science and applications.[10] Recently, it has been demonstrated that magnetic order can be changed and stabilized up to room temperature by substituting Cr by Mn.[9,11–16] Stable $(\text{Cr}_{1-x}\text{Mn}_x)_2\text{AC}$ ($A = \text{Al}$, Ge or Ga) type MAX phase compounds can be considered as a new family of magnetic MAX phases, which exhibit FM properties. Furthermore, the Mn_2GaC magnetic compound has been synthesized as a heteroepitaxial thin film containing Mn as the exclusive M element.[10,17,18]

The magnetic MAX phases have very large saturation fields (larger than 5 T) and a low ratio of remanent-to-saturation magnetization.[15–18] The magnetization of these systems increases rapidly with decreasing temperature and usually does not show the characteristics of a conventional ferromagnet.[15–20] These findings have been interpreted to be the result of competing FM and AFM correlations,[10] which can lead to non-collinear or ferrimagnetic spin structures. The crystal structure of MAX phases is hexagonal with a repeated M-A-M-X-M-A-M-X atomic layer stacking in the c direction. First-principle calculations have shown that Cr-C networks (M-X-M) favor FM spin alignment in the Cr_2GeC

*Corresponding author. Email: ruslan.salikhov@uni-due.de

system, whereas Ge (in Cr-Ge-Cr bonds) supports AFM super-exchange like coupling between Cr atoms due to the mixing of the Cr 3d and Ge 4p states.[4] However, experiments show that the electron density of pure Cr_2GeC is not spin-polarized and only Mn doping leads to FM long-range order.[9] The spin structure of new Mn-doped magnetic MAX phases is currently under investigation. It is worth noting that the sign of exchange coupling and thus the ground state (AFM or FM) can be tuned by changing the interlayer distances, which makes these systems extremely interesting for magnetocaloric and spintronic applications.[10]

Due to the hexagonal crystal structure and the reduced dimensional (quasi 2D) layer structure, the spin-orbit coupling of Mn (Cr) 3d electrons can strongly influence the magnetic properties and spin structure. Magnetocrystalline anisotropy energy (MAE) is a qualitative measure of spin-orbit interaction and can be used to estimate anisotropic orbital magnetic moments.[21] A powerful method to quantify the magnetic anisotropy energy density is ferromagnetic resonance (FMR).[22] Here, we present the results of FMR measurements on 34 nm thick $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ films epitaxially grown on $\text{MgO}(111)$ substrates. The selected MAX phase has to date the largest saturation magnetization among the magnetic MAX phase series and shows a FM response near room temperature.[16] The synthesis and structural properties of these films have been described elsewhere.[16] X-ray diffraction and cross-section transmission electron microscopy of

these samples reveal a high crystal quality with a composition of $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$. The films are phase-pure with a lateral grain size larger than 150 nm. The c -axis aligns parallel to the film normal.

Temperature- and angular-dependent FMR measurements were performed using a conventional FMR X-band setup at a microwave frequency $f = 9.23$ GHz. FMR spectra were recorded for different orientations of the static magnetic field with respect to the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ film plane (out-of-plane geometry) and within the film basal plane (called in-plane geometry from this point forward) at temperatures between 110 and 300 K. Additionally, magnetic hysteresis loops at various temperatures were measured in both geometries using SQUID magnetometry.

FMR spectra of the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ film measured at different temperatures (T) are presented in Figure 1 for in-plane (a) and out-of-plane (b) geometries. From the spectra the resonance field (H_r) was determined using the first derivative of the Lorentz function fitted to the experimental data. At $T = 105$ K the resonance field (H_r) is 282 ± 2 mT for in-plane and 436 ± 2 mT for out-of-plane geometry. This indicates that the easy axis of magnetization (M) is in the film plane.[22] With increasing T and decreasing magnetization, the resonance fields for in- and out-of-plane geometry shifts toward the same $\mu_0 H_r = 330$ mT (at $T \approx 250$ K). One can also see that with increasing temperature, the integral intensity (I) of the resonance line for both configurations decreases (Figure 1(c), black squares for in-plane

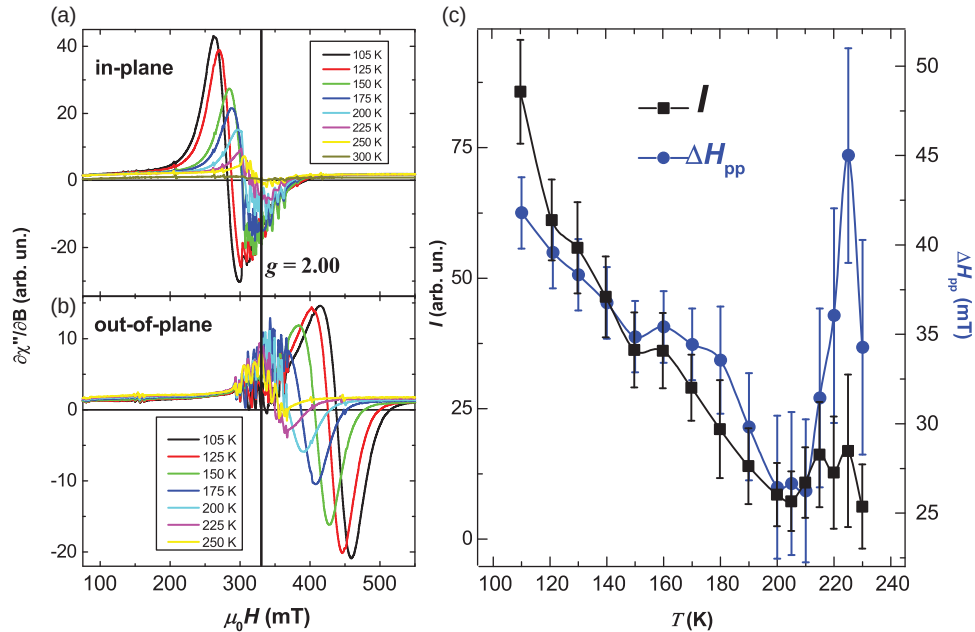


Figure 1. FMR spectra for different temperatures measured at $f = 9.2265(1)$ GHz with the magnetic field applied parallel (a) or perpendicular (b) to the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ film plane. The ‘noisy’ features seen at 300–350 mT are paramagnetic resonance (EPR) signals from impurities in the MgO substrate. (c) Temperature dependence of the FMR integral intensity I (black squares and left scale) and peak-to-peak resonance linewidth ΔH_{pp} (blue circles and right scale), solid lines are guide to the eye.

geometry) up to 205 K representing the decrease in the magnetization.[23] The FMR peak-to-peak linewidth (ΔH_{pp}) at different temperatures (blue circles in Figure 1(c)) decreases with increasing temperature similar to the FMR integral intensity, indicating that magnetic relaxation processes in the FM ($\text{Cr}_{0.5}\text{Mn}_{0.5}$)₂GaC film are slowing down with the magnitude of M . At $T > 205$ K the intensity remains unchanged within the experimental error, but ΔH_{pp} rapidly increases to peak at 225 K. This is attributed to the crossover of the magnetic relaxation rates from critical speeding up just above the Curie temperature ($T > T_C$) to thermodynamical slowing down close to T_C , as has been previously reported for the metallic thin films near an FM to paramagnetic phase transition (see, for example, Li et al. [24] and Platow et al. [25] and references therein). A residual microwave absorption signal is seen up to 300 K. We also found that at temperatures above 240 K, the resonance fields for in- and out-of-plane geometry are identical within the error bar of 3 mT. This indicates that the magnetic anisotropy above $T = 240$ K becomes very small (or zero). The above findings allow the calculation of the spectroscopic g -factor using the equation $H_r = \omega/\gamma$, where $\omega = 2\pi f$, and $\gamma = g\mu_B/\hbar$ is the electron gyromagnetic ratio.[22] We arrive at $g = 2.00 \pm 0.01$, which suggests pure spin magnetism in the ($\text{Cr}_{0.5}\text{Mn}_{0.5}$)₂GaC film. The absence of an orbital contribution to the magnetic moment usually results in a small MAE. Indeed, the in-plane resonance field is independent of the azimuthal angle (not shown) at 110, 150 and 250 K within the error bar of 0.1 mT. Since the samples are epitaxial and textured with a lateral grain size larger than 150 nm,[16] we would expect that in-plane magnetic anisotropies would be reflected in the angular dependence of the resonance field.

In the following we discuss the *out-of-plane* angular dependence of the FMR field using the Smit-Beljers approach.[22,26,27] Considering that there is no significant magnetocrystalline anisotropy in the film plane as indicated by the lack of an in-plane FMR angular dependence in the epitaxial film, we calculate the resonance frequency using the effective uniaxial anisotropy field given by the shape anisotropy $(1/2)\mu_0(N_{\perp} - N_{\parallel})M_{\text{eff}}^2$ for thin films, where M_{eff} is the effective magnetization. Note that M_{eff} (from FMR) and magnetization, M , measured by SQUID magnetometry can differ and such variations can be attributed to an out-of-plane anisotropy along the c axis. We use the demagnetization factors $N_{\perp} = 1$, $N_{\parallel} = 0$ of an infinitely thin film, which is appropriate for the respective thickness of our film.[28] The corresponding free energy (F) including Zeeman energy reads

$$F = \frac{1}{2}\mu_0 M_{\text{eff}}^2 \cos^2 \theta - \mu_0 M H (\cos \theta \cos \theta_H + \sin(\varphi - \varphi_H) \sin \theta \sin \theta_H) \quad (1)$$

with (θ, φ) and (θ_H, φ_H) being the polar (accounted from the film plane normal) and azimuthal (in-plane) angles of the magnetization M and the applied magnetic field H , respectively. The resonance frequency can then be calculated analytically using the double derivatives of the magnetic free energy F with respect to polar and azimuthal angles [22,26,27]:

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{(M \sin \theta)^2} (F_{\theta\theta} F_{\varphi\varphi} - F_{\theta\varphi}^2). \quad (2)$$

For FM films without significant in-plane MAE azimuthal angles in Equation (1) can be set to $\varphi = \varphi_H = 0$. The angles θ correspond to the equilibrium angle of the magnetization and can be determined from the condition $dF/d\theta = 0$ for each field angle θ_H at the corresponding resonance field $H = H_r$. Thus, for the out-of-plane angular dependence of resonance fields, the g -factor ($\gamma = g\mu_B/\hbar$) and M_{eff} are fitting parameters. In the simple case assuming that $\theta = \theta_H$ one finds from Equation (2) the frequency-field dispersions, known as Kittel equations:

$$\left(\frac{\omega}{\gamma}\right)^2 = \mu_0^2 H_{r\parallel} (H_{r\parallel} + M_{\text{eff}}), \quad (3)$$

when the external magnetic field is applied parallel to the film plane ($\theta = \theta_H = 90^\circ$), and

$$\left(\frac{\omega}{\gamma}\right)^2 = \mu_0 (H_{r\perp} - M_{\text{eff}}) \quad (4)$$

for the out-of-plane configuration ($\theta = \theta_H = 0^\circ$). $H_{r\parallel}$ and $H_{r\perp}$ are resonance fields detected at $\theta_H = 90^\circ$ and 0° , respectively. The values of $\mu_0 M_{\text{eff}}$ calculated using Equations (3) and (4) for two configurations (with $g = 2$) are plotted in Figure 2. One can see that at temperatures between 125 and 200 K, $\mu_0 M_{\text{eff}}$ for the in-plane configuration is smaller than for the out-of-plane. This means that the effective magnetic anisotropy at these temperatures for the out-of-plane geometry is larger than for the in-plane geometry.

To quantify the effective magnetic anisotropy more precisely, we performed angular-dependent out-of-plane measurements at 110 K from in-plane ($\theta_H = 90^\circ$) to out-of-plane ($\theta_H = 0^\circ$) orientation of the external field with respect to the film plane (Figure 3). The Smit-Beljers approach described above was used as a fit to the experimental data (black circles). From the fit (solid line in Figure 3) we found $\mu_0 M_{\text{eff}} = 103 \pm 3$ mT in good agreement with the values in Figure 2 determined using Equations (3) and (4). The spectroscopic splitting $g = 1.99 \pm 0.01$ is within the error bar identical to the one determined at ambient temperature. The $\mu_0 M$ ($4\pi M$ in cgs units) at $\mu_0 H = 290$ mT measured by SQUID magnetometry in in-plane geometry at 110 K is

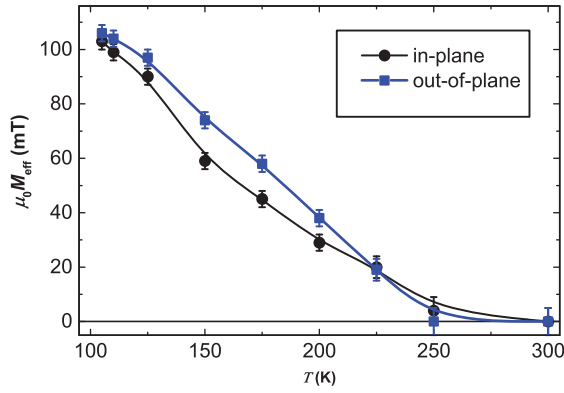


Figure 2. Effective magnetic anisotropy fields in the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ film calculated using Equations (3) and (4) for in- and out-of-plane configurations at different temperatures. Solid lines are guide to the eye.

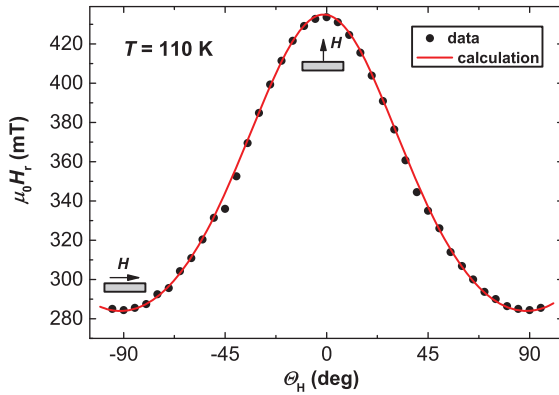


Figure 3. FMR field measured at 110 K (black circles, experimental error is approx. the dot size) as a function of angle between the applied external magnetic field and the axis normal to the film plane. The red solid line represents the result of fit using the Smit-Beljers approach with fit parameters: $\mu_0 M_{\text{eff}} = 103 \pm 3$ mT, and $g = 1.99 \pm 0.01$.

about 100 mT. This value is close to $\mu_0 M_{\text{eff}}$ at the same temperature, which leads us to following conclusions:

- (1) the magnetocrystalline anisotropy along the c axis (sample normal) is negligibly small in the $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ MAX phase system and
- (2) the Mn (Cr) spin moments are FM coupled and aligned parallel in a basal plane causing the shape anisotropy.

The possible AFM alignment of some magnetic moments in the basal plane would significantly reduce the FM long-range order, which in turn would reduce the magnetic shape anisotropy of an infinitely thin film ($N_{\perp} = 1$, $N_{\parallel} = 0$) down to the shape anisotropy of confined areas of locally FM coupled spins and for that case $\mu_0 M_{\text{eff}}$ is expected to be significantly

smaller than $\mu_0 M$. Considering that the magnetocrystalline anisotropy is negligible one can conclude that the difference between $\mu_0 M_{\text{eff}}$ for in-plane and out-of-plane configurations (Figure 2) at the temperatures between 125 and 200 K is purely related to shape anisotropy.

According to Petruhins et al.,[16] epitaxial $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ films show a magnetic moment of $0.19 \mu_B$ per M atom in a field of 5 T at 300 K and a remanent moment of $0.004 \mu_B$ per M atom. This field-induced magnetization is also seen in microwave absorption as the signal above 205 K, which stays constant up to 300 K. The temperature dependencies of I , ΔH_{pp} and $\mu_0 M_{\text{eff}}$ in Figures 1(c) and 2 point toward a ferro- to paramagnetic phase transition at $T = 205\text{--}230$ K. The peak in $\Delta H_{\text{pp}}(T)$ at 225 K (above the ordering temperature) confirms the ferro- to paramagnetic phase transition, since is characteristic for critical fluctuations of the magnetization, which have also been observed at the ferro- to paramagnetic phase transition of Ni [24], for example. All aforementioned facts support the existence of FM order in the present samples. Based on the microwave data, however, it cannot be concluded unambiguously that itinerant ferromagnetism is encountered as has been concluded before for $(\text{Cr}_{1-x}\text{Mn}_x)_2\text{GeC}$ MAX phases.[9] The difference in shape anisotropy $\mu_0 M_{\text{eff}}$ between in- and out-of-plane configurations at temperatures between 125 and 200 K in Figure 2 indicates the anisotropy in FM order of magnetic moments. This anisotropy possibly can be explained considering the nanolaminated $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ system as a quasi 2D ferromagnet, where interlayer magnetic coupling supports 3D long-range ordering at finite temperatures.[29] In the out-of-plane configuration a larger magnetization appears due to enhanced interlayer coupling when changing the orientation of magnetic moments from parallel in the basal plane to perpendicular. The magnetometry data (not shown) support our present findings; however, the origin of the anisotropy of interlayer coupling remains open.

In conclusion, temperature- and angular-dependent FMR measurements of the new magnetic $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ MAX phase reveal that the g -factor is 2.00 ± 0.01 at ambient (300 K) and low temperatures (110 K). This suggests a small (or zero) orbital contribution to the magnetic moment. The in-plane and out-of-plane magnetocrystalline anisotropies are negligible. Furthermore, we provide evidence that magnetic moments in the basal plane of nanolaminated $(\text{Cr}_{0.5}\text{Mn}_{0.5})_2\text{GaC}$ are FM coupled and this magnetic material can most likely be classified as a quasi 2D laminar ferromagnet, where interlayer coupling depends on the magnetic field direction.

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